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IMPROVED ETMOD MODULES FOR HTO EXCHANGE, HT DEPOSITION, AND VEGETATION HTO

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ABSTRACT

Ontario Hydro's environmental tritium model, ETMOD, calculates the potential radiological impact of short duration releases of HT and HTO. This report develops and documents improved modules for the following key parameters: the deposition velocity of HT to soil, the exchange velocities of HTO to soil and vegetation, and the atmospheric stability class. These new modules allow the HT deposition velocity and the HTO exchange velocities to be estimated from readily available information (eg, soil and weather conditions), ensure that these parameters are consistent with stability class and other model input (eg, wind speed, season, and time of day), allow diurnal and seasonal effects to be modelled, and reduce the need for site-specific input. In addition, a module to calculate HTO concentrations in vegetation is provided which will allow the calculation of potential ingestion dose.

job	file	date	report no.
35-140	837.262	September 10, 1991	91-191-K

CFFTP-G-91-88

1.0 INTRODUCTION

The Environmental Tritium Model (ETMOD) is a computer code which calculates the behaviour and fate of short duration atmospheric releases of tritiated hydrogen, HT, and tritiated water, HTO, for the assessment of potential radiological impact.

ETMOD is the latest in a series of codes implemented by Ontario Hydro that includes TRITMOD (Murphy and Prendergast 1979) and OHTDC (Bell et al. 1985, Russell and Ogram 1988). Like these earlier codes, ETMOD is capable of simulating the following key processes: atmospheric dispersion of the release plume; deposition of HT and HTO to soils; oxidation of HT in surface soils; subsequent re-emission of HTO to the atmosphere and its atmospheric dispersal; and HTO migration in soil.

The purpose of this report is to develop and document the following improvements to ETMOD:

(i) modules which calculate the HTO exchange velocities between air and ground surfaces (vegetation or soil) and the deposition velocity of HT to soil on the basis of weather conditions, season and land use;

(ii) a module for HTO concentration in vegetation water; and

(iii) a methodology to estimate stability class from readily available meteorological input.

These improvements extend the capability of ETMOD in the following ways:

- The new modules determine HT deposition velocity, HTO exchange velocity, and atmospheric stability class from readily available meteorological and site information. This approach allows ETMOD to treat diurnal and seasonal effects under a wide range of conditions. Use of these methods also ensures that parameters input to ETMOD such as wind speed, atmospheric stability and HTO exchange velocity are consistent. In previous versions of the ETMOD code, single values of these parameters were input independently thereby allowing the possibility of unphysical combinations. For example, stable conditions are unlikely during daylight hours and are thus unlikely to occur at the same time as a high rate of HTO exchange with vegetation, which peaks during daylight hours.

- The vegetation module will allow the time history of HTO in vegetation to be calculated and thus allow the prediction of ingestion dose.

Accurate treatment of the movement of tritium between the atmosphere, soil and vegetation is particularly important in predicting the radiological impact of HT releases (Murphy et al. 1982, Bunnenberg et al. 1990). For example, the dominant process which converts HT to the much more hazardous form of HTO is oxidation of HT by microorganisms present in soil. The HTO thus formed is incorporated into soil moisture from which it can be taken up by vegetation and/or

re-emitted to the atmosphere, thereby giving rise to potential ingestion, immersion and inhalation doses. Re-emission of HTO from soil and vegetation to the atmosphere occurs during a period of days to weeks after the release. As a result, the dose impact during the passage of the HT plume may be far less than the potential impact occurring in the period after plume passage due to the gradual re-emission of HTO.

The new methods are described in the following sections. The HT deposition module is based largely on a simple expression for the resistance to HT uptake by soil given by Dunstall and Ogram (1990). Multiple resistance methods developed recently for regional scale acidic deposition and oxidant formation models have been adapted to treat the exchange of HTO between vegetation and soil moisture and the atmosphere. The method of estimating stability class is based on a method implemented for nuclear emergency response in Ontario (Wright 1986) and is similar to a method recommended by the Ontario Ministry of the Environment in its draft Air Pollution Regulation 308 (OME 1987). The vegetation module is that recommended in a recent review of key processes and parameters for environmental tritium models (Bunnenberg et al. 1990). These improvements have recently been encoded in ETMOD by S.B. Russell of the Nuclear Safety Department.

The advantages of these methods are illustrated by means of a specific example using weather conditions measured during the HT dispersion experiment conducted at Chalk River in June 1987.

2.0 HT DEPOSITION

Model sensitivity studies have shown that the dominant process controlling the impact of HT releases is the oxidation of HT in surface soils (Raskob 1990, Murata 1990). This oxidation process causes a net flux of HT from the atmosphere to the soil. The deposition flux is modelled in ETMOD by a deposition velocity defined as the deposition flux divided by the concentration of HT in air at a reference height, usually taken to be 1 m above the surface. Because of its importance, the deposition velocity has been the subject of a number of experimental studies in the laboratory and field.

The HT deposition module is based on the multiple resistance approach shown schematically in Figure 1. The deposition velocity is expressed as the reciprocal of the sum of three resistances as shown in equation (1): the resistance to aerodynamic transport from the reference height to the surface, R_a; the resistance to transport through the laminar sublayer adjacent to the surface, R_b; and the resistance to uptake at the surface, R_c.

$$v_g^{-1} = R_a + R_b + R_c \qquad (1)$$

The resistance components R_a, R_b and R_c can be estimated from input meteorology and readily available information such as soil bulk density and moisture content as follows.

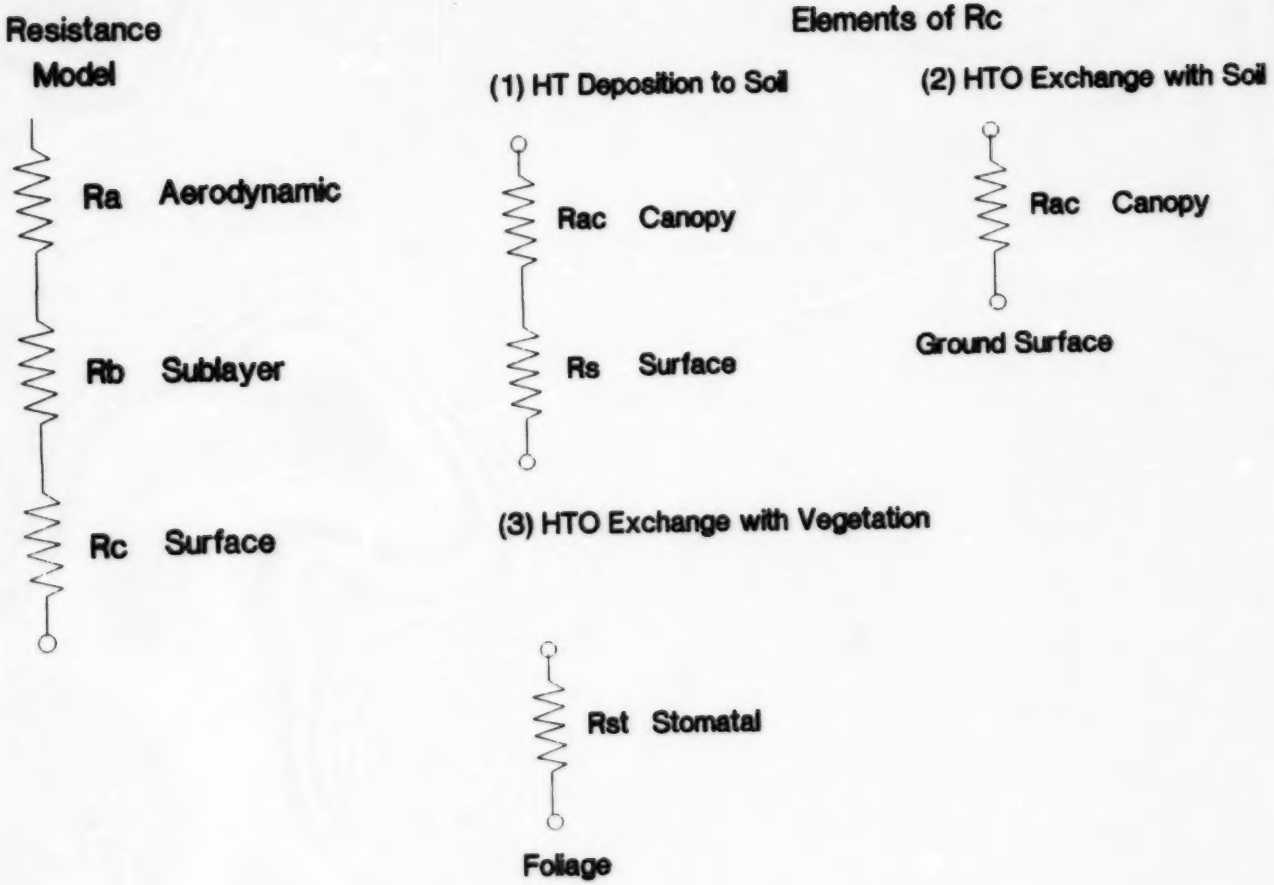

Fig. 1 Multiple resistance models for HT deposition and for HTO exchange.

Widely accepted methods (Hicks et al. 1987) are adapted here to calculate the resistances R_a and R_b for HT deposition (and for HTO exchange, as will be discussed later in Section 3). These methods are summarized in Table 1.

The resistance to turbulent atmospheric transport, R_a, is independent of the species of gas being considered and is a function of reference height (for species concentration), surface roughness length, z_0, friction velocity, u_*, and atmospheric stability, the latter characterized by Monin-Obukhov length, L. Roughness lengths given by Wesely and Lesht (1988) for various land use types as a function of season are listed in Appendix A. The determination of u_* and L is discussed below in Section 5.

The resistance to diffusive transport through the quasi-laminar sub-layer, R_b, depends on the gas species and can be estimated by the expression given in Table 1. This expression was developed by examining deposition data obtained over a variety of surfaces (Hicks et al. 1987).

As shown in Figure 1, the surface resistance, R_c, for HT deposition to soil is comprised of two components: R_s, the resistance to uptake at the soil surface; and, R_{sc}, the resistance to transport through the vegetation canopy, if any (direct uptake of HT by vegetation is slow enough to be neglected). The surface uptake resistance R_s is estimated from an approximate expression derived by studying deposition to samples of bare soil taken from eleven Ontario sites and prepared with various water contents (Dunstall and Ogram 1990). This study found that, in most cases, the HTO depth profile resulting from HT deposition could be described adequately by an exponentially decreasing profile with scaling depth, z_s, and that the deposition velocity could be approximated by a simple expression in terms of an average z_s of 0.023 m and the effective HT diffusion coefficient in soil, D_{eff}, ie:

$$R_s = z_s / D_{eff} \qquad (2)$$

where $D_{eff} = D_o \theta_a / \tau$. D_o is the gas phase diffusion coefficient of HT, θ_a is the air filled volume fraction (estimated from soil bulk density and water content, and an assumed soil particle density of 2.65×10^3 kg m^{-3}), and τ is a tortuosity factor (assumed to be 1.5). D_o (HT) at 20°C is approximately 0.51×10^{-4} m^2 s^{-1}.

This expression for R_s is expected to be most accurate under biologically active (non-freezing) conditions with moderate soil water content. Under extreme conditions (for example, high soil water content, or below freezing or very high temperatures) the above expression would underestimate R_s, and hence overestimate the deposition velocity. This is a conservative approach as the rate of conversion of the HT release to HTO, and hence its radiological impact, is nearly proportional to the deposition velocity (Ogram 1984). For HT deposition, it is worth noting that R_s is usually greater than R_a, R_b or R_{sc} so that $v_g \sim R_s^{-1}$ (Dunstall et al. 1985).

TABLE 1

Components of HTO Exchange Velocity and HT Deposition Velocity

Aerodynamic Resistance:

$$R_a = (1/ku_*)\ [\ln((z_r-d)/z_0) - \phi(z_r/L)]$$

Sublayer Resistance:

$$R_b = (2/ku_*)\ (Sc/Pr)^{2/3}$$
$$R_b(HT) = 2.9/u_*$$
$$R_b(HTO) = 5.0/u_*$$

Surface Resistance: ***1. HT deposition to soil***

$$R_c = R_s + R_{ac}$$

where

$$R_s = 0.023/D_{eff}$$

2. HTO exchange with soil

$$R_c = R_{ac}$$

3. HTO exchange with vegetation

$$R_c = R_{st}$$

$$R_{st} = R_{min}\{1+[200(G+0.1)^{-1}]^2\}\{400[T_s(40-T_s)]^{-1}\}$$

Symbols:

D_{eff} [$m^2\ s^{-1}$]:	*effective diffusion coefficient for HT in soil*	
d [m]:	*displacement height (~0.5 z_0)*	
G [$W\ m^{-2}$]:	*incoming solar radiation*	*(See Appendix D)*
k:	*Von Karman's constant (0.35)*	
L [m]:	*Monin-Obukhov length*	*(See Appendix D)*
Pr:	*Prandtl number*	
R_a [$m\ s^{-1}$]:	*aerodynamic resistance*	
R_b [$m\ s^{-1}$]:	*laminar sub-layer resistance*	
R_c [$m\ s^{-1}$]:	*bulk surface resistance*	
R_{ac} [$m\ s^{-1}$]:	*resistance to canopy transport*	*(See Appendix B)*
R_{st} [$m\ s^{-1}$]:	*bulk canopy stomatal resistance*	*(See Appendix C)*
R_s [$m\ s^{-1}$]:	*resistance to soil HT uptake*	
Sc:	*Schmidt number*	
T_s [°C]:	*surface air temperature.*	
u_* [$m\ s^{-1}$]:	*friction velocity*	*(See Appendix D)*
z_0 [m]:	*surface roughness length*	*(See Appendix A)*
z_r [m]:	*reference height (1 m)*	
Φ:	*stability correction factor*	*(See Appendix D)*

Values of R_{ac} determined on the basis of land use and season are given by Wesely (1989). A table of these values is given in Appendix B.

The HT deposition module is shown schematically in Figure 2.

3.0 HTO EXCHANGE VELOCITY

Exchange of HTO occurs between the moisture in air and the water in terrestrial surfaces such as vegetation (see below), soil, snow, and dew. In ETMOD the exchange flux is modelled as the product of an exchange velocity, v_{ex}, and the difference between the HTO vapour pressure in air and the equilibrium vapour pressure of HTO in the surface component under consideration.

In previous versions of the ETMOD code a single value for the exchange velocity between vegetation and soil was used. The exchange velocity is, however, a function of atmospheric conditions, season, time of day and the nature of the surface.

To account for this dependence, the exchange velocity to soil, $v_{ex,soil}$ and the exchange velocity to vegetation, $v_{ex,veg}$, like the deposition velocity of HT to soil, can be determined from a multiple resistance approach as shown in Figure 1. R_a and R_b are calculated from the expressions given in Table 1. In the case of vegetation, R_c depends on the bulk canopy resistance to stomatal exchange, R_{st}, (expressed per unit ground area). For exchange with soil moisture, R_c is given by the resistance, R_{ac}, to transport through any ground cover (such as a vegetation canopy).

Tables of minimum values of R_{st} for water vapour (H_2O) exchange as a function of land use type and season have recently been developed for regional-scale acidic deposition modelling (Wesely 1989). These values should also apply to HTO because of the small differences between the diffusivities of these two isotopic forms of water. The minimum value of R_{st} is multiplied by a correction factor (Wesely 1989) which to account for variations in R_{st} with incoming solar radiation and air temperature.

It is convenient to make the simplifying assumption that HTO exchange between the atmosphere and vegetation foliage and between the atmosphere and the ground surface can be modelled independently even though, in fact, the sum of the deposition fluxes to soil and vegetation is limited by the aerodynamic and sub-layer resistances. This latter constraint can be neglected if $R_a \geq R_a + R_b$. Even in the cases where this inequality does not hold, modelling soil and vegetation deposition fluxes independently is conservative in the sense that the total HTO exchange flux is overestimated.

Direct movement of tritium between vegetation and soil via the canopy atmosphere is also implicity neglected by treating the soil and vegetation exchange fluxes independently. This latter assumption appears consistent with the results of field experiments (eg, Ogram 1988). For bare soil, snow or water surfaces, or vegetated surfaces under non-biologically active conditions (late fall or winter) the module sets R_{st} to a very large value and exchange with the ground surface will dominate.

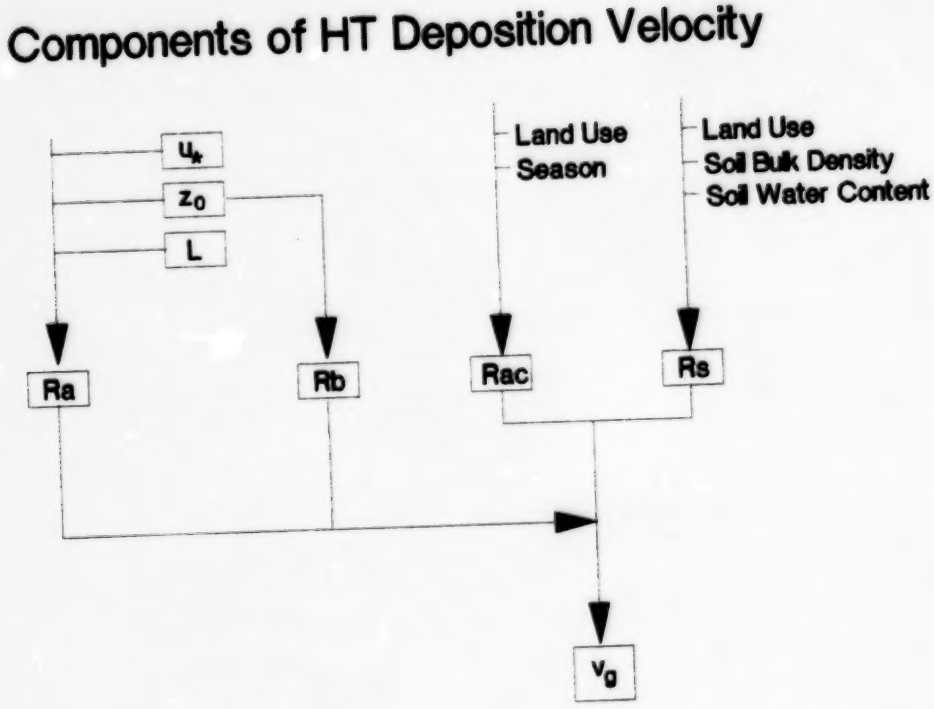

Fig. 2 Schematic of the HT deposition module.

The HTO exchange velocity module is shown schematically in Figure 3.

4.0 VEGETATION MODULE

The new ETMOD module describing tritium dynamics in vegetation treats movement of tritium between vegetation and the atmosphere (deposition and emission) as an HTO exchange process, and accounts for incorporation of HTO into vegetation by uptake via transpiration of contaminated soil water (Bunnenberg et al. 1990).

$$\frac{d}{dt}(C^{pw}_{HTO}) = v_{ex,veg}\,(C^{a}_{HTO} - \gamma h\, C^{pw}_{HTO})\,/M_{w} + T_{e} C^{srz}_{HTO}\,/M_{w} \qquad (3)$$

where: C is HTO concentration per unit mass of plant water (pw), per unit mass of water in the soil root zone (srz), and per unit volume of air (a) at the reference height, respectively; h is the saturation humidity at leaf temperature (assumed to be equal to air temperature); and M_w is the mass of vegetation water per unit ground surface area. γ is the ratio of the vapour pressures of HTO and H_2O. T_e is the flux of water from the surface due to transpiration.

$$T_{e} = v_{ex,veg}\,(h - h^{a}) \qquad (4)$$

where h^a is the ambient humidity.

5.0 FRICTION VELOCITY, MONIN-OBUKHOV LENGTH AND ATMOSPHERIC STABILITY

In order to determine the aerodynamic and sublayer resistances, R_a and R_b, it is necessary to calculate the Monin-Obukhov length, L, and friction velocity, u_*. L and u_* are well defined parameters that describe the degree of thermally and mechanically induced turbulence in the surface layer of the atmosphere (eg, Lyons and Scott, 1990). L and u_* are calculated from readily available meteorological and site information through iteration of couple equations for these two parameters (see Appendix D).

The incoming solar radiation, G, (which is also a necessary input for R_a) can be determined from time of day, date, latitude, longitude and cloud cover (Wright 1986, OME 1987). The Monin-Obukhov length, L, is calculated from its definition for daytime hours. L depends on the surface heat flux, H_0, which can estimated from G by an empirical relation (Wright 1986, OME 1987). For nighttime, L is estimated from a correlation given by Venkatram (1980).

The atmospheric stability class can be inferred from L and the surface roughness length, z_0, by a correlation developed by Golder (Golder 1972, Lyons and Scott 1990).

The atmospheric stability class module is shown schematically in Figure 4. Details of the module are given in Appendix D. Linking the determination of atmospheric stability class to the determination of HTO exchange velocity and HT deposition velocity ensures that these model inputs are consistent with each other and with the assumed time of day, season and land use type.

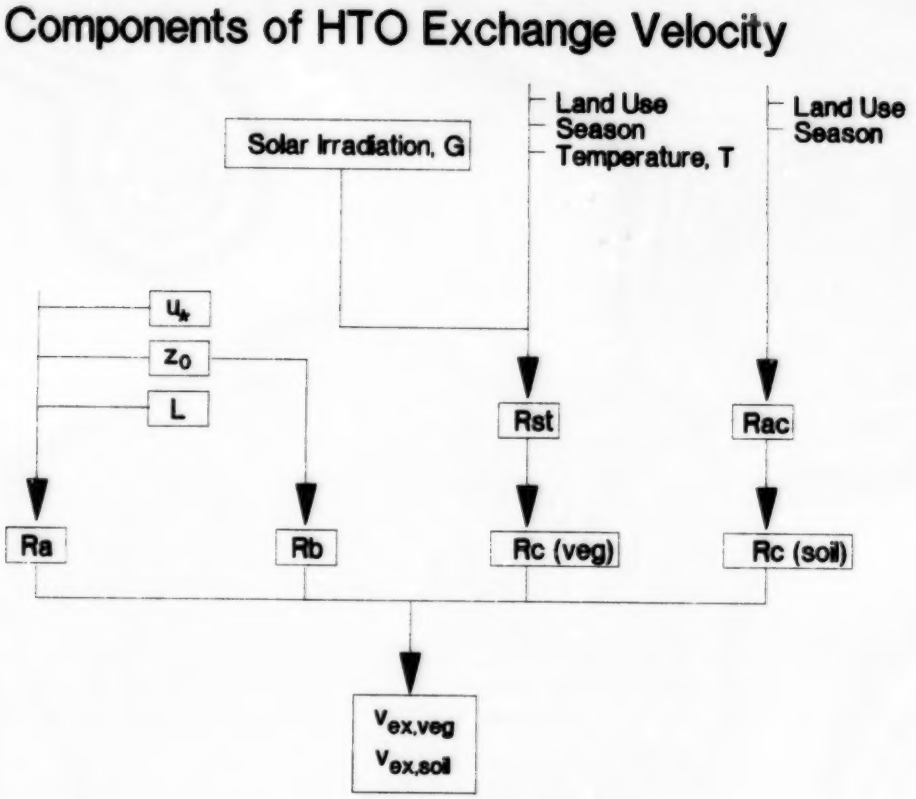

Fig. 3 Schematic of the HTO exchange module.

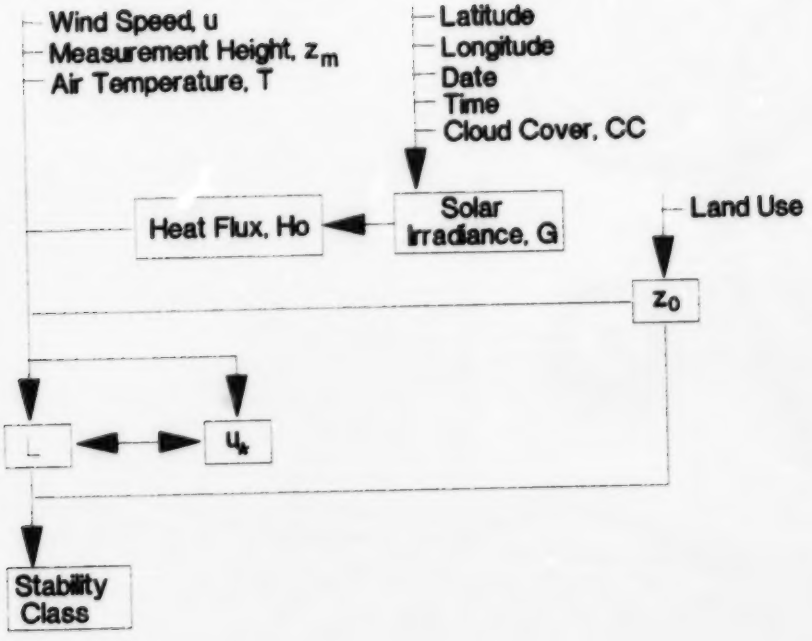

Fig. 4 Schematic of the atmospheric stability class module.

6.0 EXAMPLE OF USE OF HTO EXCHANGE AND STABILITY CLASS MODULES

Use of the formulas given in this report is illustrated by calculating atmospheric stability class and HTO exchange velocities to soil and vegetation for a three day period (June 15 to 17, inclusive) following the June 10, 1987, HT dispersion experiment at Chalk River (Burnham et al. 1988). The wind speed and cloud cover were determined from on-site meteorological measurements (Brown et al. 1988).

Figure 5 shows the measured wind speed at 2.8 m height together with stability class and HTO exchange velocities to soil and vegetation as determined by the recommended formulas. The figure shows clearly the diurnal variation in exchange velocities and stability class and the correlation of these parameters with wind speed.

7.0 CONCLUDING REMARKS

The ETMOD code predicts the behaviour of short-duration releases of HT and HTO. This report provides new modules which allow the code to calculate key input parameters such as atmospheric stability class, soil HT deposition velocity, and soil and vegetation HTO exchange velocities on the basis of readily available meteorological and site information (such as site latitude and longitude, wind speed, land use type, season, time of day, soil bulk density and soil water content). Advantages of these new modules are that they (i) ensure that derived parameters are consistent with other model input, (ii) allow diurnal and seasonal effects to be modelled and (iii) reduce the need for site-specific input. In addition, a module to calculate HTO concentrations in vegetation is provided which will allow the prediction of potential ingestion dose.

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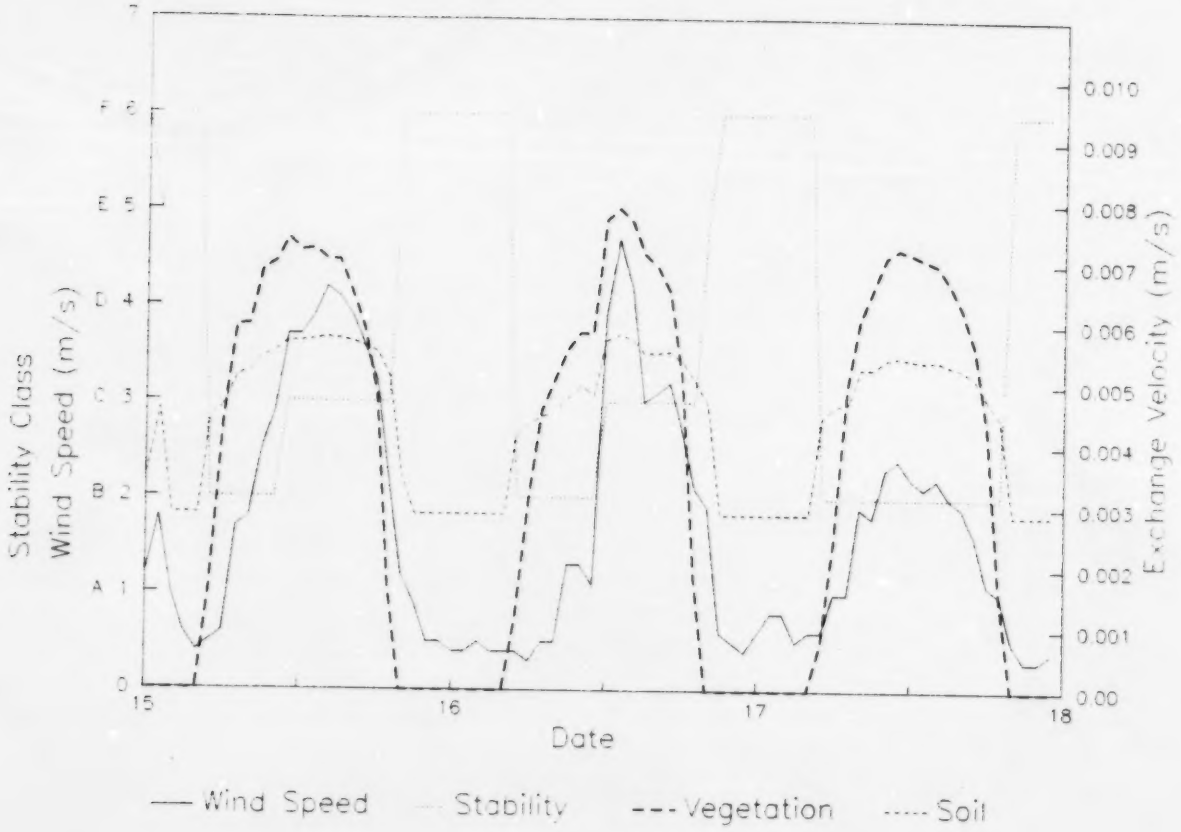

Fig. 5 Example of calculated atmospheric stability classes and HTO exchange velocities for soil and vegetation at the site of the June 1987, Chalk River HT dispersion experiment for the three day period, June 15 to 17, 1987, inclusive.

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APPENDIX A

Table A1 gives surface roughness length for various land use types by season.

TABLE A1

Land Use Type	Surface Roughness Length, z_0 (m) Spring	Summer	Fall	Winter
1 Agricultural	0.03	0.25	0.1	0.001
2 Range	0.02	0.05	0.05	0.001
3 Mixed Agric./Range	0.03	0.1	0.08	0.001
4 Deciduous Forest	1	1	1	1
5 Conifer Forest	1	1	1	1
6 Mixed Forest	1	1	1	1
7 Barren (Desert)	0.002	0.002	0.002	0.002
8 Rocky, Open, Low Shrubs	0.06	0.1	0.08	0.04
9 Urban	1	1	1	1
10 Water	0.001	0.001	0.001	0.001

NOTE: Values for land use types 1 to 9 as recommended by Wesely and Lesht (1989).

APPENDIX B

Table B1 gives the resistance to transport through the vegetation canopy to the ground surface for various land use types by season.

TABLE B1

Land Use Type	Resistance to Transport Through Canopy to Soil, R_{ac} (s m^{-1}) Spring	Summer	Fall	Winter
1 Agricultural	50	200	150	10
2 Range	80	100	100	10
3 Mixed Agric./Range	60	150	120	10
4 Deciduous Forest	1200	2000	1000	1000
5 Conifer Forest	2000	2000	2000	2000
6 Mixed Forest	1500	2000	1500	1500
7 Barren (Desert)	0	0	0	0
8 Rocky, Open, Low Shrubs	120	200	140	50
9 Urban	100	100	100	100
10 Water	0	0	0	0

NOTE: Adapted from Wesely (1989).

APPENDIX C

Table C1 gives the minimum bulk canopy resistance to water vapour (H_2O) exchange for various land use types by season.

TABLE C1

Land Use Type	Minimum H_2O Bulk Canopy Resistance, R_{min} (s m^{-1}) Spring	Summer	Fall	Winter
1 Agricultural	120	60	*	*
2 Range	240	120	*	*
3 Mixed Agric./Range	200	100	*	*
4 Deciduous Forest	140	70	*	*
5 Conifer Forest	250	130	250	400
6 Mixed Forest	190	100	500	800
7 Barren (Desert)	-	-	-	-
8 Rocky, Open, Low Shrubs	300	150	*	*
9 Urban	-	-	-	-
10 Water	-	-	-	-

NOTE 1: Adapted from Wesely (1989).
NOTE 2:
* Indicates Large Resistance
"-" Indicates Not Applicable

APPENDIX D

Derivation of u_* and L

Average values of the Monin-Obukhov length, L, and the friction velocity, u_*, can be estimated from readily available meteorological data by the methods used in Ontario's nuclear emergency response model (Wright 1986) and recommended by the Ontario Ministry of the Environment in its Draft Air Pollution Regulation 308 (OME 1987). These methods are described below.

(1) For hours between sunrise and sunset, the surface heat flux, H_0, is first estimated as

$$H_0 = 0.35G - H_L \qquad (1)$$

where G is the incoming solar radiation and H_L is the long wave heat loss from the surface. G is given by:

$$G = 950\ \beta\ (\cos(\phi)\cos(\tau)\cos(L_R) + \sin(\phi)\sin(L_R))\ \text{W m}^{-2} \qquad (2)$$

where:

$\tau = \pi\ (t-t_n)/12$;
t is time (taken as half way through hour);
$t_n = 12 + L_D/15$ is local noon;
$\phi = \tan^{-1}\ (0.438 \sin \pi((d-78)/180))$;
d is Julian day (1 to 365);
L_D is longitude (degrees);
L_R is latitude (radians); and
β is radiation reduction factor for cloud cover, cc, from Table D1.

TABLE D1

Cloud Cover, *cc* (%)	β
0	1
12.5	0.89
25	0.81
37.5	0.76
50	0.72
62.5	0.67
75	0.59
87.5	0.45
100	0.23

H_L is given by:

$$H_L = -3\ (8.55 - cc)\ \text{W m}^{-2}$$

where cc is the cloud cover in eighths.

(2) If H_0 is greater than 5 W m^{-2} the boundary layer is assumed to be convective (unstable) and L and u_* are calculated by iteration from the definition of L, ie:

$$L = -u_*^3\ \bar{T}\ \rho\ c_p\ /\ (0.35\ g\ H_0) \qquad (3)$$

where:

$\bar{T}$ is mean boundary layer temperature (K);
ρ = 1.2 kg m^{-3} is density of air;
c_p = 1005 J kg^{-1} K^{-1} is heat capacity of air at constant pressure;
g = 9.81 $m^2 s^{-1}$ is the gravitational acceleration; and
H_0 is the surface heat flux (W m^{-2});

and the relation

$$u_* = 0.35\ u\ /\ (ln(z_a/z_0) - \Phi(z_a/L)) \qquad (4)$$

where:

$\Phi(z_a/L) = 2\ ln((1+q)/2) + ln((1+q^2)/2) - 2\ arctan\ q + \pi/2$;
$q = (1-15z_a/L)^{0.25}$;
u is wind speed (m s^{-1});
z_a is wind measurement height (m); and
z_0 is roughness length.

(3) If H_0 is less than 5 W m^{-2} during daytime or if the ground is snow covered, the atmosphere is assumed to be neutral, ie, 1/L ~ 0.

(4) Between sunrise and sunset, u_* and L are determined by iteration from the correlations given by Venkatram (1980).

$$u_* = 0.35\ u/(ln(z\Phi(za/L)) \qquad (5)$$

where:

$$(\Phi z_a/L) = -\ 4.7(z_a/L)$$

$$L = 1100\ u_*^2 \qquad (6)$$

(5) Time of sunrise and time of sunset is determined by the following relations.

$$t_{rise} = t_n - (12/\pi)cos^{-1}\ (-tan(\phi)tan(L_R)) \qquad (7)$$

$$t_{set} = t_n + (12/\pi)cos^{-1}\ (-tan(\phi)tan(L_R)) \qquad (8)$$

(6) Stability class is derived from L and z_0 by Golder's correlation (Golder 1972, Lyons and Scott 1990) which is shown in Figure D1.

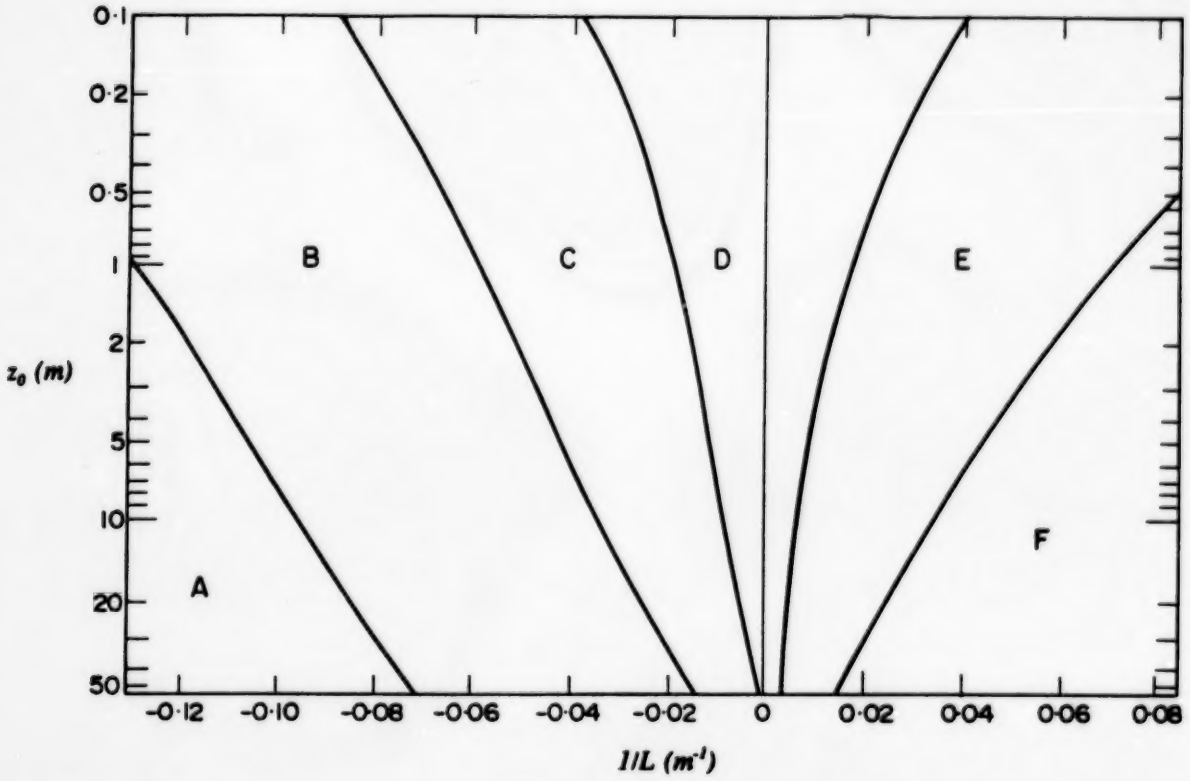

Fig. D1 Relationship between atmospheric stability class, Monin-Obukhov length, L, and surface roughness length, z_0 (Golder 1972).